

This Page Is Inserted by IFW Operations  
and is not a part of the Official Record

## **BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning documents *will not* correct images,  
please do not report the images to the  
Image Problem Mailbox.**





1712  
image

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Application of

Atty. Docket No.

YTSEN WIELSTRA ET AL

NL010052

Serial No.: 10/047,024

Group Art Unit: 1712

Filed: 15 JANUARY 2001

Ex.: D.S. METZMAIER

METHOD OF PREPARING A LACQUER COMPOSITION

Commissioner for Patents  
Alexandria, VA 22313-1450

CLAIM FOR PRIORITY

Sir:

In response to the Examiner's request in paragraph 2 of his Detailed Action dated December 19, 2002, enclosed please find a Certified copy of the European Application No. 01200191.3 filed January 18, 2001.

Respectfully submitted,

By *Ernestine C. Bartlett*  
Ernestine C. Bartlett, #22,861  
Attorney  
(914) 333-9640

Enclosure

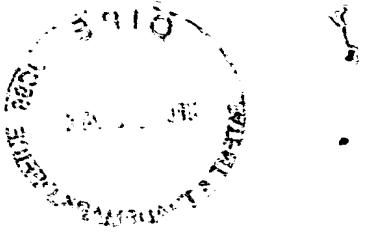
CERTIFICATE OF MAILING

It is hereby certified that this correspondence is being deposited with the United States Postal Service as first-class mail in an envelope addressed to:

COMMISSIONER FOR PATENTS  
Alexandria, VA 22313-1450

On 12/17/2003

By G. Lamparski





**Europäisches  
Patentamt**

**European  
Patent Office**

**Office européen  
des brevets**

**Bescheinigung**

**Certificate**

**Attestation**

Die angehefteten Unterlagen stimmen mit der ursprünglich eingereichten Fassung der auf dem nächsten Blatt bezeichneten europäischen Patentanmeldung überein.

The attached documents are exact copies of the European patent application described on the following page, as originally filed.

Les documents fixés à cette attestation sont conformes à la version initialement déposée de la demande de brevet européen spécifiée à la page suivante.

**Patentanmeldung Nr. Patent application No. Demande de brevet n°**

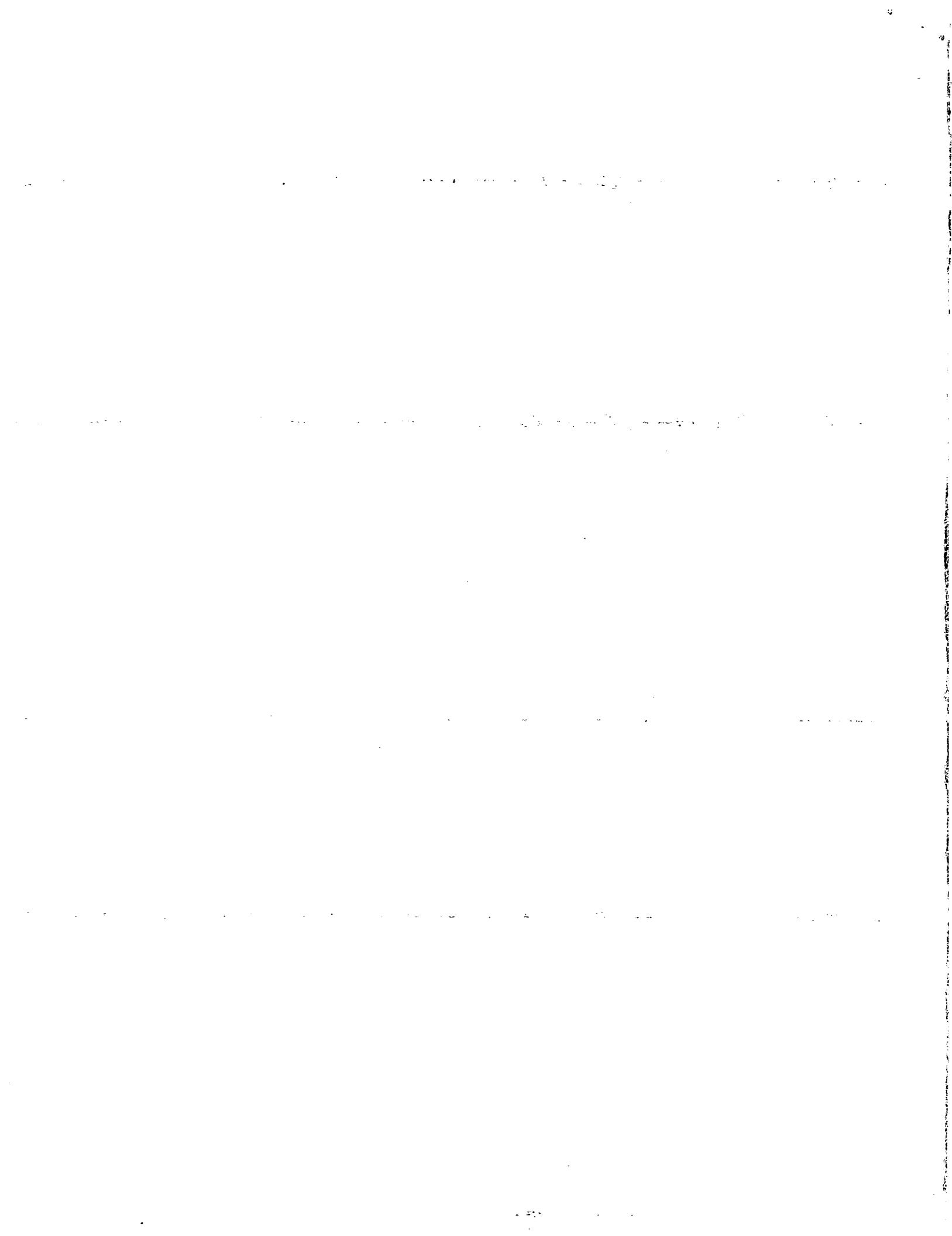
01200191.3

Der Präsident des Europäischen Patentamts;  
Im Auftrag

For the President of the European Patent Office

Le Président de l'Office européen des brevets  
p.o.

I.L.C. HATTEN-HECKMAN





Anmeldung Nr:  
Application no.: 01200191.3  
Demande no:

Anmelde tag:  
Date of filing: 18.01.01  
Date de dépôt:

Anmelder/Applicant(s)/Demandeur(s):

Koninklijke Philips Electronics N.V.  
Groenewoudseweg 1  
5621 BA Eindhoven  
PAYS-BAS

Bezeichnung der Erfindung/Title of the invention/Titre de l'invention:  
(Falls die Bezeichnung der Erfindung nicht angegeben ist, siehe Beschreibung.  
If no title is shown please refer to the description.  
Si aucun titre n'est indiqué se referer à la description.)

Method of preparing a lacquer composition

In Anspruch genommene Priorität(en) / Priority(ies) claimed /Priorité(s)  
revendiquée(s)

Staat/Tag/Aktenzeichen/State>Date/File no./Pays/Date/Numéro de dépôt:

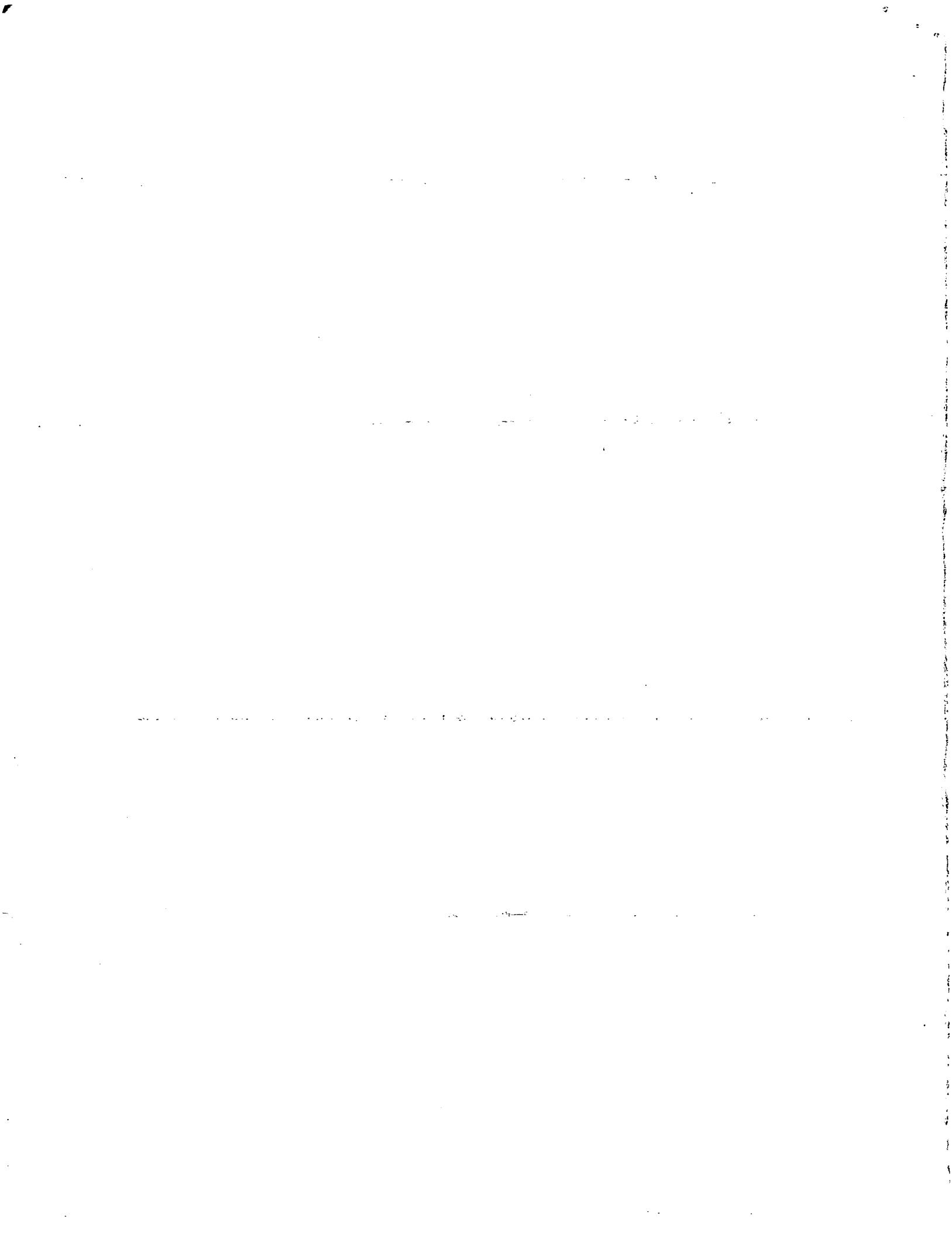
/00.00.00/

Internationale Patentklassifikation/International Patent Classification/  
Classification internationale des brevets:

C09D4/00

Am Anmelde tag benannte Vertragsstaaten/Contracting states designated at date of  
filing/Etats contractants désignées lors du dépôt:

AT BE CH CY DE DK ES FI FR GB GR IE IT LI LU MC NL PT SE TR



PHNL010052EPP

**Method of preparing a lacquer composition**

The present invention relates to a method of preparing a lacquer composition, said method comprising the steps of mixing together an organosilane compound and silica particles under basic conditions. The invention also relates to a lacquer composition which is obtainable by the method according to the invention, as well as to a method of applying a lacquer coating to a substrate.

The lacquer compositions of the above mentioned type, also referred to as sol-gel materials, are usually made by hydrolysis of silanes forming Si-OH groups, which are coupled towards Si-O-Si bonds upon curing. Silanes can be hydrolyzed both in an acidic environment as well as in a basic environment. However, the behavior of the Si-OH groups in an acidic environment differs from the behavior of said groups in a basic environment. The different behavior of the Si-OH groups in basic and acidic conditions is, inter alia, disclosed in "Sol-Gel Science; The Physics and Chemistry of Sol-Gel Processing", chapter 2, by C.,J. Brinker and G.W Scherer.

Under acidic conditions, coupling of the Si-OH groups is relatively slow. This slow coupling results in a positive effect on the potlife of a lacquer made under the same conditions. Upon curing of the material, a forced coupling takes place forming chain-like structures which resemble normal organic polymers.

However, under basic conditions, coupling of the Si-OH groups is relatively fast, due to the high reactivity of the Si-OH groups. As a consequence of said fast coupling of Si-OH groups particles of nanometer size are formed, instead of chain-like structures. This process is hard to control and can result in flocculation and gelation. Upon curing of the coating the layers turn white.

The present invention aims to provide for a method of preparing a lacquer composition under basic conditions, which method provides for a controlled coupling of Si-OH groups and which method prevents premature gelation.

To this end, the present invention provides for a method according to the preamble, which method is characterized in that a metal alkoxide is added to the reaction mixture.

PHNL010052EPP

By adding a metal alkoxide to the basic reaction mixture the coupling reaction can be suppressed. As the metal complex reacts with SiOH forming metal-OSi bonds an uncontrolled growth is prevented. An important advantage of the method according to the present invention is that new materials can be made with different properties.

5 The basic conditions under which the lacquer composition is prepared, are preferably caused by the addition of ammonia to the reaction mixture.

The benefit of the method according to the present invention especially applies for lacquer compositions to which silica particles are added in order to provide for increased strength and reduced shrinkage. As the iso-electric point of silica is 2, and therefore the silica 10 particles are electrically neutral at pH 2, a poor stabilization of silica particles is obtained at said pH. Under more basic conditions the particles are negatively charged resulting in improved stability and reduced tendency towards flocculation. Therefore, upon curing of such basic lacquer improved packing of the particles is obtained with better properties of the final coating.

15 It is important that the metal alkoxide reacts with SiOH and not with water. Therefore, the metal alkoxide preferably comprises a zirconium alkoxide or an aluminum alkoxide or a mixture thereof.

Preferably, the metal alkoxide comprises a diketonate, such as metal ethylacetacetate or a metal acetylacetone or a mixture thereof.

20 Although the organosilane compound may comprise different reactive groups, it is preferred to use an epoxysilane, preferably 3-glycidyloxypropyltrimethoxysilane (GLYMO).

In order to influence the properties of the network of the cured lacquer coating, advantageously at least a second organosilane compound is present.

25 In particular the mechanical strength of the cured lacquer layer can be improved by using a tetra-alkoxysilane, such as tetra-alkylorthosilicate (TEOS) or tetra-methylorthosilicate (TMOS).

The properties of the lacquer composition can further be improved by the addition of organic compounds, such as epoxides and polyalcohols or by the addition of 30 inorganic materials, such as metaloxides, like aluminumoxide or titaniumoxide.

The present invention further relates to a lacquer composition which is obtainable by the method according to the present invention as disclosed in the above.

Said lacquer composition has excellent film-forming properties. Moreover, the lacquer composition may be sprayed on the substrate to form a lacquer coating, if necessary

PHNL010052EPP

by adding a diluting agent. The lacquer coating which is obtained by curing the lacquer composition according to the present invention is transparent and meets high quality standards from an optical and an esthetical point of view. Moreover, the lacquer coating proves to be wear-resistant and scratch-resistant, even after prolonged exposure to solvents or 5 compositions for personal care, such as shaving lotions. This makes the application of the lacquer useful in electric shavers, such as the shaver head or a housing.

Furthermore, the present invention relates to a method of applying a lacquer coating to a substrate in which method a lacquer composition is applied to the substrate and cured, thereby forming the lacquer coating. Said method is characterized in that a lacquer 10 composition according to the present invention is used.

The lacquer composition can be provided by means of methods known per se, such as spraying, spin-coating, curtain-coating or dip-coating. The lacquer composition can be cured by means of heating. The appropriate temperature and the duration of the thermal treatment are governed by the lacquer composition used and can be determined in a manner 15 known to the skilled person, for example by measuring the hardness of the lacquer coating.

Finally, the invention relates to a product which is provided with a lacquer coating, said product being characterized in that the lacquer coating is obtained by using the method of applying a lacquer coating to a substrate according to the present invention.

The lacquer composition according to the invention can be applied to a great 20 variety of products. The lacquer can have both the function of a protective coating as well as improving the attractive properties of the product provided with the lacquer. Examples of products which may be provided with the lacquer are housings for electr(on)ical products, such as television receivers, personal digital assistants, mobile phones, and personal computers; personal-care products, such as shavers; domestic appliances, such as irons, and 25 luminaires.

The invention will be further elucidated by means of the following examples.

30 Examples 1-3: Preparation of the metal alkoxides

Example 1

A quantity of 17.4 g ethylacetooacetate was added to 63 g Zr(OC<sub>3</sub>H<sub>7</sub>)<sub>4</sub> (70% in propanol) at room temperature and the solution was stirred. The resulting solution in which

PHNL010052EPP

4

18.01.2001

ethylacetacetate has bonded to the zirconium is called zirconium ethylacetacetate (ZrEAA). When the amount of ethylacetacetate was doubled Zr(EAA)<sub>2</sub> was formed.

#### Example 2

5 A quantity of 13.4 g acetylacetone was added to 63 g Zr(OC<sub>3</sub>H<sub>7</sub>)<sub>4</sub> (70% in propanol) at room temperature and the solution was stirred. Zirconium acetylacetone (Zr(AcAc)) was formed. When the amount of acetylacetone was doubled Zr(AcAc)<sub>2</sub> was formed.

#### 10 Example 3

A quantity of 40 g Al(OC<sub>4</sub>H<sub>9</sub>)<sub>3</sub> was added to 21 g ethylacetacetate at room temperature and the solution was stirred. This resulted in the formation of a solution of aluminum ethylacetacetate (AIEAA).

#### 15 Examples 4-9: Preparation of the lacquer compositions

#### Example 4

Half of a mixture of 19.6 g glycidyloxypropyltrimethoxysilane (GLYMO), 8.6 g tetra-ethylorthosilicate (TEOS) and 30 g alcohol was added to 44 g of an aqueous suspension of silica particles (Ludox AS40, Dupont de Nemours), while the solution was stirred. After 2 minutes the other half of the mixture was added. After stirring during 7 minutes, 7.8 g AIEAA, prepared according to the method as disclosed in example 3, was added. Subsequently, 1.4 g ammonia (25%) was added to the solution. The pH was measured to be 7.36.

25 The mixture was observed for hydrolysis of the silanes. Hydrolysis is said to be complete when the composition gives proper wetting on a glass substrate.

After 4 days hydrolysis of the silanes was not completed yet.

#### Example 5

30 Half of a mixture of 19.6 g GLYMO, 8.6 g TEOS and 30 g alcohol was added to 44 g Ludox AS40, while the solution was stirred. After 2 minutes the other half of the mixture was added. After stirring during 7 minutes, 7.8 g AIEAA, prepared according to the method as disclosed in example 3, was added. Subsequently, 2.8 g ammonia (25%) was added to the solution. The pH was measured to be 9.5.

PHNL010052EPP

Hydrolysis of the silanes was completed within 4 hours. After curing of the lacquer a transparent wear-resistant layer was obtained.

#### Example 6

5 Half of a mixture of 23.7 g GLYMO, 5.2 g TEOS and 30 g alcohol was added to 44 g Ludox AS40, while the solution was stirred. After 2 minutes the other half of the mixture was added. After stirring during 7 minutes, 11.8 g ZrAcAc, prepared according to the method as disclosed in example 2, was added. Subsequently, 1.4 g ammonia (25%) was added to the solution. The pH was measured to be 9.0.

10 Hydrolysis of the silanes was completed within 4 hours. After curing of the lacquer a transparent wear-resistant layer was obtained.

#### Example 7

15 Half of a mixture of 19.6 g GLYMO, 8.6 g TEOS and 30 g alcohol was added to 44 g Ludox AS40, while the solution was stirred. After 2 minutes the other half of the mixture was added. After stirring during 7 minutes, a mixture of 9.3 g ZrEAA and 3.9 g AlEAA, prepared according to the methods as disclosed in example 1 and 3 respectively, was added. Subsequently, 2.8 g ammonia (25%) was added to the solution. The pH was measured to be 10.0.

20 Hydrolysis of the silanes was completed within 1 hour. After curing of the lacquer a transparent wear-resistant layer was obtained.

#### Example 8

25 A mixture of 19.6 g GLYMO and 30 g ethanol was added to 44 g Ludox AS-40 while the solution was stirred. After 2 minutes 6.3 g tetra-methyl orthosilicate (TMOS) was added. After stirring during 7 minutes, a mixture of 9.3 g ZrEAA and 3.9 g AlEAA was added. The pH was measured to be 7.8.

Hydrolysis of the silanes was complete after 15 minutes. After curing of the lacquer a transparent and wear-resistant layer was obtained.

30

#### Example 9 (comparative example)

Half of a mixture of 19.6 g GLYMO, 8.6 g TEOS and 30 g alcohol was added to 44 g Ludox AS40, while the solution was stirred. After 2 minutes the other half of the

PHNL010052EPP

6

18.01.2001

mixture was added. Subsequently, 2.8 g ammonia (25%) was added to the solution. The pH was measured to be 10.5.

Hydrolysis of the silanes was completed within 1 hour. After curing of the lacquer a cloudy, brittle layer was obtained.

5

Example 10: Testing of the lacquer, obtained by applying the compositions of examples 5-8 on a substrate and curing said lacquer compositions

10 A quantity of 3 g aluminum pigment (E2140 of Silberline) was added to each of the lacquer compositions prepared according to examples 5, 6, 7 and 9, respectively, and dispersed under stirring. The lacquer compositions obtained were sprayed on a substrate of IXEF 1032 (Solvay polyarylamide 60%GF), resulting in a metallic appearance of the substrate. Thereupon, the coatings were cured at 160°C during 20 minutes. A coating  
15 thickness of 20 micron was obtained.

The chemical resistances of the lacquers were tested by immersing the coated substrate in commercial shaving lotions during 28 days. The lacquer coatings passed said test without being attacked or softened.

20 The mechanical resistances of the lacquers were tested by bringing the lacquer coatings into contact with a rotating disc-shaped abrasive sponge (Scotch Brite), with a compressive force of 500 g during 20 minutes. After said treatment, no signs of wear were observed.

PHNL010052EPP

## CLAIMS:

1. A method of preparing a lacquer composition, said method comprising the steps of mixing together an organosilane compound and silica particles under basic conditions, characterized in that a metal alkoxide is added to the reaction mixture.
- 5 2. A method according to claim 1, characterized in that the metal alkoxide is a zirconium alkoxide or an aluminum alkoxide or a mixture thereof.
- 10 3. A method according to claim 1 or 2, characterized in that the metal alkoxide comprises a metal diketonate.
4. A method according to claim 1, characterized in that the organosilane compound is an epoxysilane.
- 15 5. A method according to claim 4, characterized in that the epoxysilane is 3-glycidyloxypropyltrimethoxysilane.
6. A method according to claim 1, characterized in that at least a second organosilane compound is present.
- 20 7. A method according to claim 6, characterized in that the second organosilane compound comprises a tetra-alkoxysilane.
8. A lacquer composition, obtainable by a method according to any one of the preceding claims.
- 25 9. A method of applying a lacquer coating to a substrate in which method a lacquer composition is applied to the substrate and cured, thereby forming the lacquer coating, characterized in that a lacquer composition as claimed in claim 8 is used.

PHNL010052EPP

10. Product provided with a lacquer coating, characterized in that the lacquer coating is obtained by using the method as claimed in claim 9.

**ABSTRACT:**

Disclosed is a method of preparing a lacquer composition, said method comprising the steps of mixing together an organosilane compound and silica particles under basic conditions. A metal alkoxide is added to the reaction mixture in order to prevent premature gelation of the reaction mixture. A lacquer composition which is obtainable by the 5 above method is also disclosed, as well as a method of applying a lacquer coating to a substrate and the coated substrate obtained.

